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STRATOSPHERIC COLUMN NO2 MEASUREMENTS FROM THREE ANTARCTIC SITES

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The significance of stratospheric odd-nitrogen compounds in Antarctic ozone depletion studies has prompted an increase in our Antarctic activities since 1986. Although several species are being studied, work has concentrated on the acquisition of NO2 data. Ground-based measurements of stratospheric column NO2 have been made at Arrival Heights, Antarctica, since spring 1982, with some gaps in the data base. Additional data has been acquired since February 1986 at Pole Station and Halley Bay, thus providing a chain of stations across the continent.

The technique used is that of absorption spectroscopy in several wavelength regions, although here we only report on those measurements in the 430-450nm region where strongly structured absorption features due to NO2 are identified in scattered sunlight in the zenith sky. Operation of a moon-tracking system at Arrival Heights has provided some additional data during the polar night. Previous analyses have shown that the NO2 column observed from the ground is strongly influenced by the season, and by the location of the site with respect to that of the polar vortex. The column amount correlates strongly with stratospheric temperature at about 70 mbar. The present data set further illustrates these features, and demonstrates both the strengths and qualifications apparent in the technique.

Data are acquired over a range of solar zenith angles around dawn and dusk twilight, and plots of data taken at 90 degrees show the seasonal trends in the observed slant column. Figure 1 shows a composite data set for Arrival Heights for 1982-1987. In general the afternoon values are higher than in the morning because of release of NO2 from its overnight storage reservoir. It can be seen that there is a steady seasonal decline in NO2 in the autumn data, and a corresponding increase in spring, caused in the main by movement of NO2 to and from the polar night storage reservoirs, thought to be N2O5 and HNO3. It should be pointed out, however, that the large 90 degree slant column values are a consequence of enhanced absorption in the long path at twilight; and model calculations show that the enhancement factor, proportional to airmass, is also dependent on layer height. Our initial calculations suggest that the bulk of the NO2 layer in autumn is probably at a height of about 20km, whereas in spring it is located much higher, perhaps 30km. If the layer is rising during autumn, therefore, some reduction in the observed slant column of up to 25 percent could result,without any real change in the vertical column. Large quasi-cyclic variations in spring are the result of observing NO2 enriched air from lower latitudes as the vortex moves away from the site. A more detailed examination of the year by year plots shows

1) Onset, at about day 60, of conversion of NO2 to N2O5 as continuous daytime photolysis of NO3 first ceases, even though the layer is still continuously, but weakly, illuminated:

2) Rather uniform behaviour in autumn, but large differences in spring as planetary wave activity changes from year to year:

3) A suggestion in the 1987 data of a reduction in the diurnal variation of NO2, which may mean that more NOx is sequestered in HNO3 at the expense of N2O5:

4) Consistently low values of NO2 in spring 1987, evidently associated with the low temperatures that prevailed.

Figure 2 shows the combined data for Halley Bay, Pole Station and Arrival Heights for 1986. Because 90 degree data can only be obtained from Pole at the equinox, the Pole values have been extrapolated to 90 degrees, and are hence subject to some uncertainty. The data clearly show the response of the stations to the particular location of the polar vortex.

Figure 3 shows an interesting data set from Pole Station acquired near the 1987 autumn equinox. During the course of day 90/91 the solar zenith angle increases by only half a degree, but the data clearly show a diurnal variation of up to twenty percent in the slant column. We interpret this as the signature of a horizontal spatial gradient in NO2 seen in the slant column as the sun moves around the station at effectively constant solar zenith angle. Some of the variation may also be due to a spatial change in the effective height of the NO2 layer.

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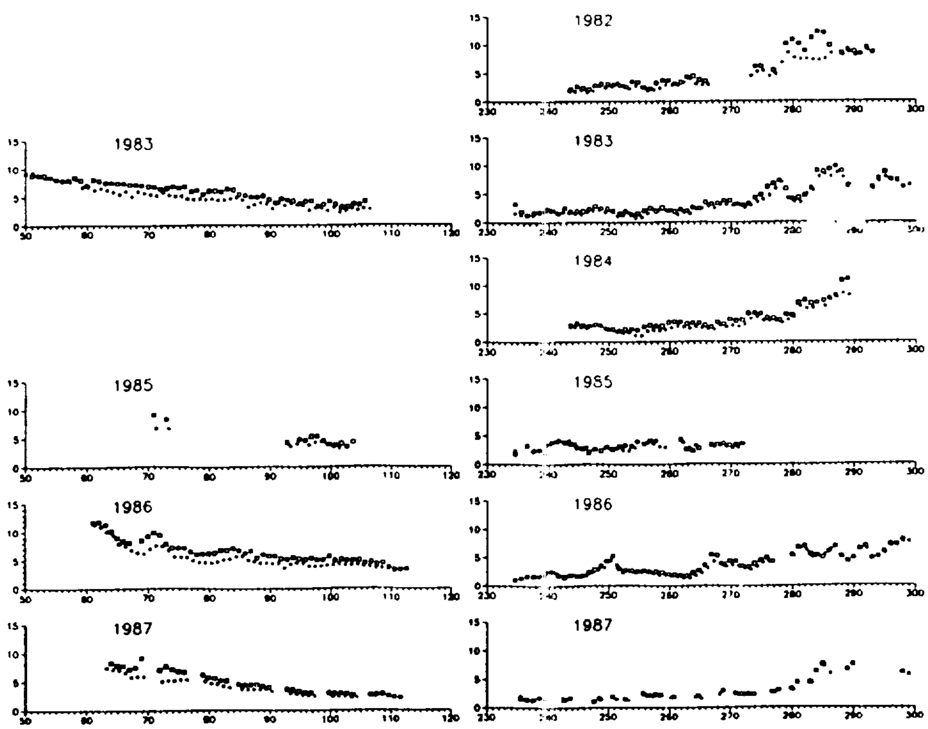

FIGURE 1: ARRIVAL HEIGHTS
SLANT COLUMN NO2(1E16 MOL CM-2)
1982-1987. AM(·), PM(•)

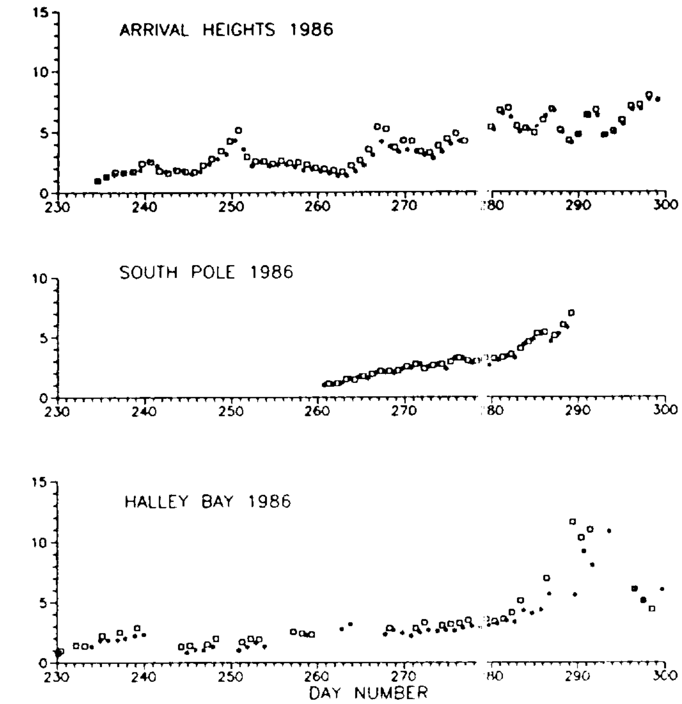

FIGURE 2: SLANT COLUMN NO2
(1E16 MOL CM-2)

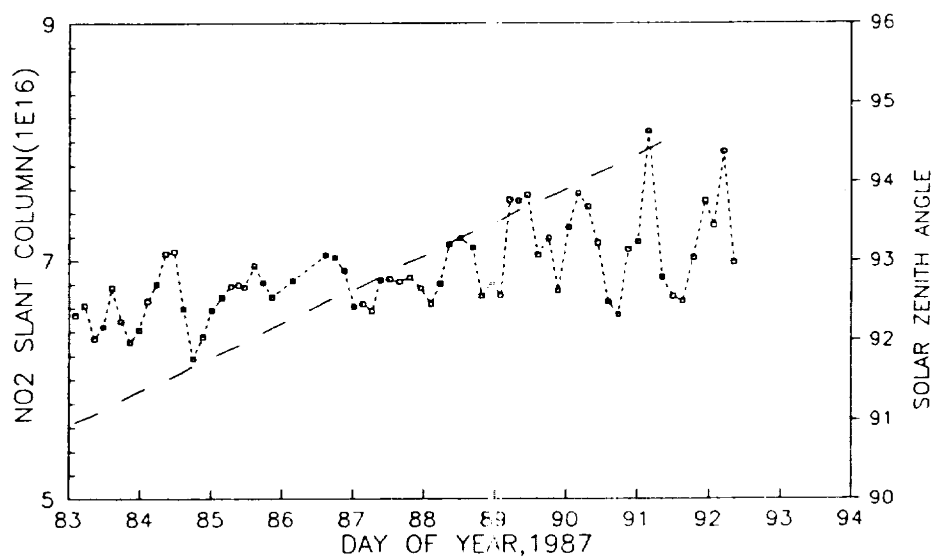

FIGURE 3: SOUTH POLE, ANTARCTICA
SLANT COLUMN NO2
3-HOUR AVERAGES